A Simple Aerosol Emissions Parameterization in RAMS
By Ted Letcher and William Cotton

1. Introduction

It has been shown through observational data and numerical experimentation that increases in CCN number concentration can have an impact on wintertime orographic snowfall. The most widely discussed physical mechanism for this is the inhibition of riming on windward mountain slopes due to smaller cloud droplets associated with high CCN concentrations (Borys et al. 2000; Saleeby et al. 2008; Saleeby et al. 2011). The impact that increased CCN has on *total* precipitation is thought to be relatively small; the increase in CCN concentration does not significantly reduce the total amount of snowfall. Instead however, the increase in CCN causes a general shift in snowfall from the upwind to the downwind side of mountain barriers (Saleeby et al. 2008). Additional numerically based studies have suggested that the effects of increased CCN on orographic snowfall are not necessarily as simple as the above mechanism may seem. Recent studies have shown that the windward precipitation decrease associated with the inhibition of riming near the mountain crest may be somewhat compensated for by the reduction of collision coalescence at lower elevations on the windward side of the mountain (Lynn et al. 2007). What the majority of these studies lack however, is a realistic representation of atmospheric CCN concentrations. Ward et al. (2011) presented a method to predict CCN concentrations in RAMS by using periodic output from the WRF-CHEM model. This method however proved highly expensive in terms of computational expense. Additionally, significant errors and biases, primarily associated with the differences between the two models, were seen using this method. The purpose of this paper is to present an alternative method to predict CCN number concentration. A method that is encompassed entirely within RAMS, therefore removing potential errors associated with different model physics, and is much more computationally efficient than the method presented by Ward et al.

2. Wintertime Sources of Cloud Nucleating Aerosol

The region immediately upwind of the Colorado Western slope is generally rural and undeveloped and as a result the air-quality along the western slope is relatively clean with, on average, lower aerosol mass concentrations compared to more urban regions in the United States (Levin and Cotton, 2009).

Climatologically, the chemical composition of the aerosol observed in this region is best described as “Remote Continental.” In the sub-micron particle range, remote continental aerosol is made up mostly of sulfate and organic compounds (Levin and Cotton, 2009). The majority of the sulfate is thought to be anthropogenic in nature, while the organic compounds are mostly from natural sources (Levin and Cotton, 2009). The sulfate aerosol (as well as an overwhelming portion of the organic aerosol) are formed as secondary aerosol, resultant from gas phase chemical processes in the atmosphere. During the winter months, primary biogenic sources of organic aerosol such as pollen and wildfire ash are likely non-existent. Similarly, secondary aerosol derived from natural sources are reduced due to reduced biogenic activity (Andrea and Rosenfeld, 2008). Numerous studies have shown that the mass fraction of organic aerosol to the total aerosol is significantly reduced during the winter months in this region (Liao et al. 2007; Schichtel et al. 2008). A brief analysis of data collected from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network revealed similar results. Fig.1 shows the monthly average aerosol mass concentration (PM2.5) measured by IMPROVE for four locations along the Colorado Western Slope. From this figure, it is seen that organic carbon

mass is lowest during the winter months, consistent with previous literature.

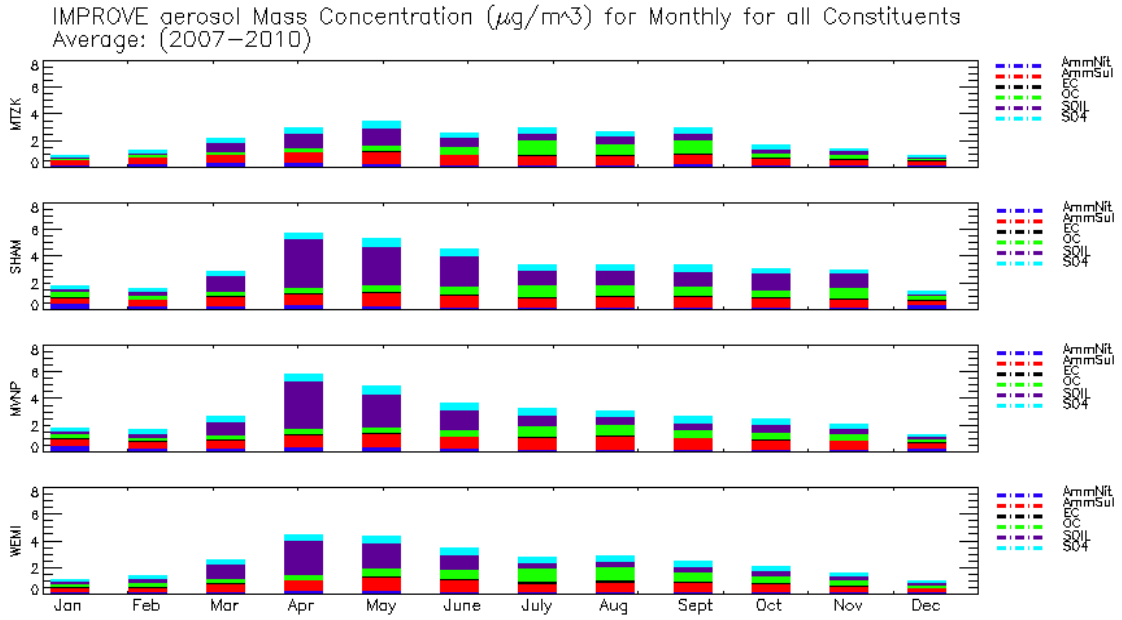

Illustration 1: Monthly averaged aerosol mass concentration [μg m^{-3}] for MTZK (40.53N, 106.69W, 3243m), SHAM (37.30N, 107.48W, 2351m), MVNP(37.19N, 108.49W,2172m) , WEMI (37.65N, 107.79W, 2750m)

The observed sulfate aerosol within the region while of secondary origin, is likely introduced into the atmosphere through combustion and the emission of gaseous sulfate compounds, most abundantly, sulfur dioxide (Levin and Cotton, 2009). These compounds undergo oxidation to form aerosol within the nucleation mode size before growing through condensation and coagulation to attain sizes more relevant to cloud nucleation. Owing to their relatively small size and abundance, these aerosols are more likely to undergo long range transport than larger aerosol that have faster settling velocities. During the winter months, the formation of new sulfate particles in the atmosphere is somewhat reduced owing to decreased photochemical activity (Levin and Cotton, 2009). As a result, much of the sulfate aerosol is likely to form in regions where the precursor gas concentrations are high, e.g., power plants and urban centers, and potentially less likely to form in more remote regions. Due to the highly reduced contribution of natural organic aerosol and the likely higher fractional amount of "near source" formation of sulfate aerosol during the winter months, it is hypothesized that, during the winter, a large portion of the total aerosol can be accounted for in a model parameterization that treats secondary aerosol as source dependent primary aerosol.

3. Aerosol Emissions Parameterization

By removing the actual chemical processes involved in forming new cloud nucleating aerosols from the emissions scheme, the computational expense drops significantly. Additionally, to simplify the budgeting of aerosol mass and number concentration, aerosol size is held constant. By making this assumption, only aerosol number concentration is required as a variable, instead of both

number *and* mass concentration. Additionally, by assuming a constant aerosol size, condensation and coagulation, processes that affect the shape of the aerosol size-distribution are not relevant. While these assumptions reduce the overall real-world representation of this scheme, they significantly cut back on computational requirements, while still providing a reasonable estimate of atmospheric aerosol concentrations for sizes relevant to cloud nucleation.

The sources used for this parameterization are derived from WRF-CHEM output. The WRF-CHEM model couples the widely used WRF model to various modules used to resolve both gas-phase and solid-phase chemical interactions in the atmosphere. This model is highly useful as it resolves the secondary formation of aerosol through gas-phase chemistry, as well as aerosol growth through condensation and coagulation. As a result, WRF-CHEM is capable of predicting both aerosol number concentration and mass concentration. Furthermore, because it resolves chemical interactions between numerous chemical species, it can provide information relating to the chemical makeup of the aerosol, and as a result, the aerosol hygroscopicity. It is because of these strengths that WRF-CHEM was chosen as a means to generate a map of aerosol sources to be used in the RAMS emissions parameterization.

To create an emissions file for RAMS, WRF-CHEM was initialized starting at 12 UTC on a day in September. The model was run at 20km resolution on a domain that covered the western 2/3rds of the United States, including the Pacific Coast. The chemistry in the model was prescribed using the National Emissions Inventory (NEI2005) data-set (EPA, 2012), and the Model Emissions of Gases and Aerosol from Nature (MEGAN). The model was run for one-hour, and the resulting near surface aerosol number concentration within the accumulation mode at each grid point was saved into a gridded file as an hourly emission rate (Fig.2 a). Additionally, aerosol hygroscopicty (represented by the symbol κ [Petters and Kreidenweis, 2007]) was determined for each source by taking the volume weighted average of each chemical species output from WRF-CHEM (Fig.2 b). The values used for each species can be found in Tab.2. from Ward et al. (2011).

Once this file was saved, the RAMS model interpolated the emission rates to match the prescribed RAMS domain and grid spacing, and then added aerosol into two model-levels surrounding a user-prescribed height. This was done instead of simply adding the aerosol to the surface layer, and served as an ad hoc representation of unresolved boundary layer mixing and secondary aerosol formation. κ was updated at each grid-point by simply taking the number concentration weighted average of the newly added aerosol and the aerosol already in place at each respective grid-point. This was appropriate, as aerosol size and density are held constant in the scheme, so a number concentration weighted average is equivalent to a volume weighted average.

To validate the accuracy of this scheme, it was used in two case-study simulations that fell within the Inhibition of Snowfall by Pollution Aerosol (ISPA) III field campaign. The ISPA-III field campaign collected CCN data at Mesa Verde National Park (MVNP) in southwest Colorado near Four Corners. This data provided the comparison data for the model simulations. Three experimental RAMS simulations were performed with varying emission heights (z_e=surface, z_e=100m, z_e=boundary layer top). In addition to the three experimental simulations, a control simulation (one without aerosol emissions) was run. Lastly, a WRF-CHEM simulation was included in the experiment to provide a model-to-model comparison for the experimental simulations. The model was run on a domain covering the western 2/3rds of the United States, at 20km horizontal resolution. The relative coarse grid-spacing was used so that the case-studies could be simulated in WRF-CHEM within a reasonable amount of time. In each case-study the model was given a 24 hour spin up period before the data collection, to give the model a chance to disperse the aerosol throughout the domain.

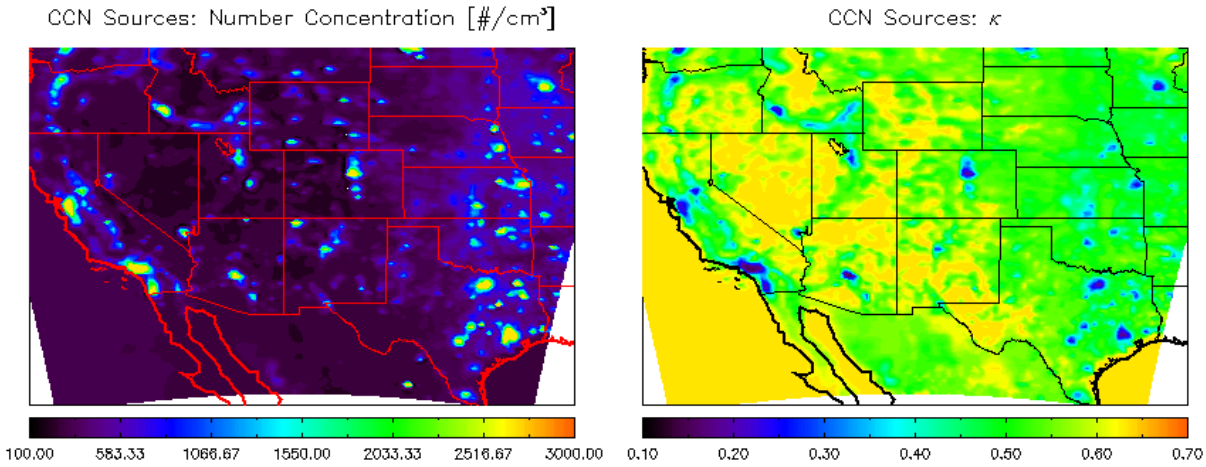

Illustration 2: a) Aerosol number concentration (potential CCN) as an hourly emission rate derived from WRF-CHEM output. b) κ as derived from the volume weight concentration of WRF-CHEM chemical species

4. Results

The first case-study simulated was of the development and passage of a synoptic trough that moved through the Four Corners region between September 28th and October 1st in 2009. The model output aerosol (potential CCN) number concentration was compared to the observed CCN (ss=3%) number concentration at MVNP. The output potential CCN from RAMS was multiplied by a factor of 0.4 to represent a mean activated fraction, making the model more comparable to the observations. The time-series of this case-study with the observations and the model simulations is shown in Fig. 3. The RAMS simulations that used the emissions parameterization performed better than the control simulation, although, in general, it exhibited a low-bias compared to the actual observations at MVNP. The simulation with z_e=100 meters performed better than the surface and the boundary layer simulations. The WRF-CHEM simulation performed reasonably throughout much of the case-study, though it produced a very large overestimate in aerosol number concentration in association with the passage of the synoptic trough. The aerosol fields produced by RAMS and WRF-CHEM appeared qualitatively similar, with the largest concentrations of aerosol east of the Rocky Mountains across the Great Plains, likely in association with the urban centers in Texas, e.g., Houston. Similarly a plume of high aerosol concentration developed ahead of the synoptic trough in both WRF-CHEM and RAMS. Although the method of aerosol scavenging used in RAMS appeared to handle the removal of aerosol better than the method used in WRF-CHEM. Similar to the first case-study, the 2nd case-study encompassed the development and passage of a synoptic trough that moved over the Four Corners region. The time-series of the observed and modeled CCN concentration is shown in Fig.4. Similar to the first case-study, the z_e=100m simulation compared the best to the observations at MVNP. Additionally, the WRF-CHEM output aerosol number concentration far exceeded reasonable values in proximity to the frontal passage. The comparison between WRF-CHEM and RAMS again showed qualitative similarities, suggesting that the emissions scheme is capable of reasonably simulating heterogeneous fields of aerosol.

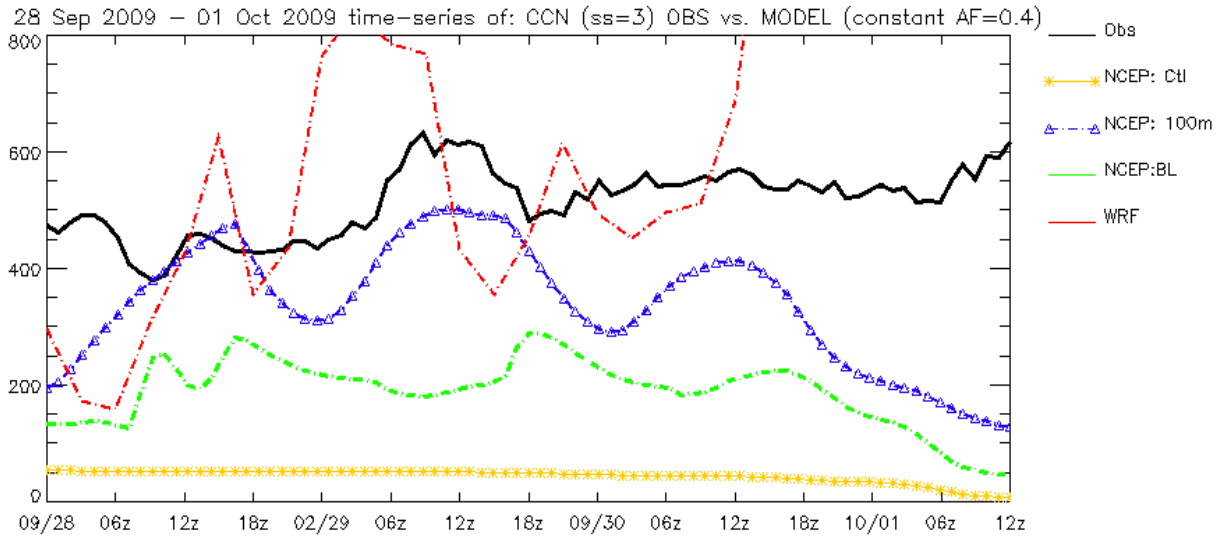

Illustration 3: RAMS output potential CCN concentration multiplied by a constant factor (0.4) and observed CCN (SS=3%) at MVNP. Time Series valid for the Sept 28 2009 – Oct 1 2009 case

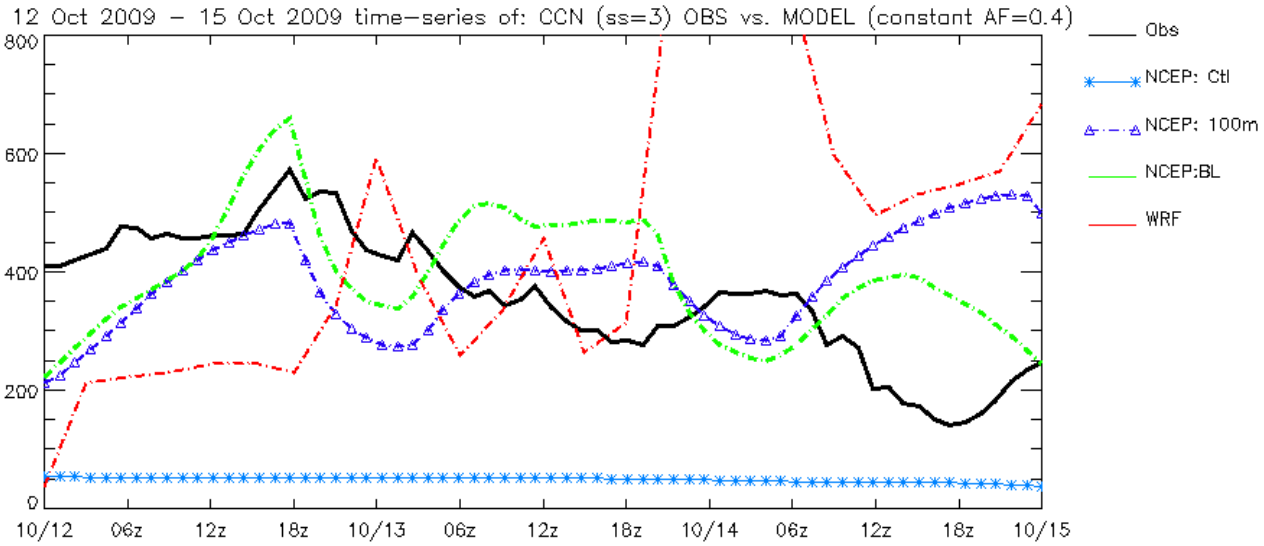

Illustration 4: RAMS output potential CCN concentration multiplied by a constant factor (0.4) and observed CCN (SS=3%) at MVNP. Time Series valid for the Oct 12 2009 – Oct 15 2009 case

5. Orographic Snow

The emissions scheme was used as part of an orographic snow simulation to improve upon the understanding of the anthropogenic influence on water resources in Colorado. The case-study performed by Saleeby et al. (2008) was chosen for repetition, as this case is well documented by both model and observations. This case-study focused on an orographic snowstorm that occurred in

northwestern Colorado on February 11th, 2007, and was measured at Storm Peak Lab (SPL) during the ISPA-II field campaign on the summit of Mt. Werner in Steamboat Springs Colorado. The model domain (shown in Fig.5) was set up identical to the domain used by Saleeby et al. with 4 two-way nested grids centered over the Park Range in northwest Colorado. The parent grid was given a horizontal resolution of 60km and covered the western 2/3rds of the United States. The smallest nested grid had a horizontal resolution of 750 meters and covered only the region in the immediate vicinity of SPL. Three simulations were run using RAMS, a control clean (CLEAN) simulation that used a background potential CCN concentration of 100 (cm^{-3}) at the surface, a control polluted (DIRTY) simulation that used a background potential CCN concentration of 1900 (cm^{-3}) at the surface, and an experimental simulation with aerosol emissions (EMISS). The EMISS simulation was also initialized with a background potential CCN concentration of 100 (cm^{-3}) at the surface. In all three simulations the vertical structure of the potential CCN number concentration was a linear decrease with height from the surface concentration to 100 (cm^{-3}) at 4km. The CLEAN and the DIRTY simulations were identical to the clean and the polluted cases simulated in Saleeby et al. (2008). All three simulations were given 24 hours of spin up time.

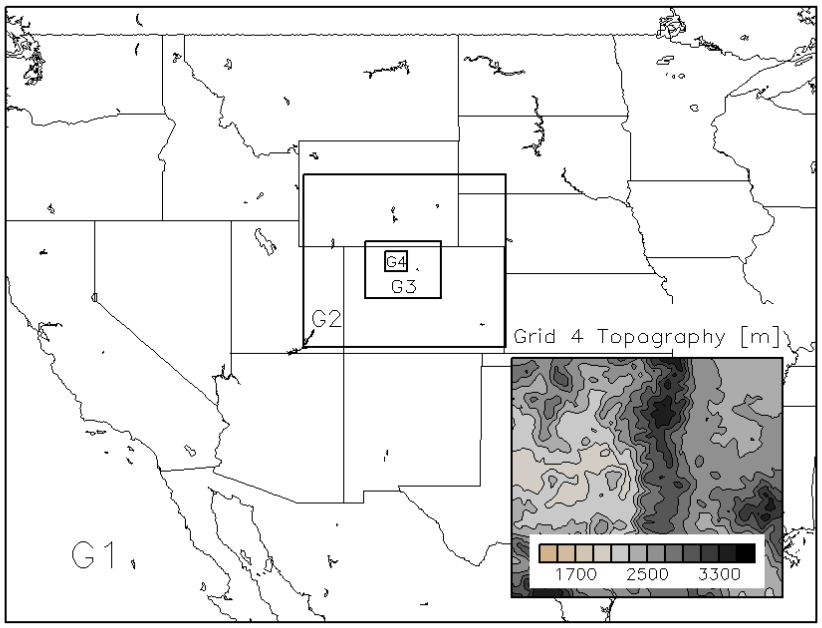

Illustration 5: Domain used to simulate the orographic snow event on Feb 11-12 2007

Results

The simulated total accumulated precipitation at the end of the CLEAN model simulation was similar enough to the Saleeby et al. clean simulation to conclude that the model performed adequately. In general, the difference in total precipitation between the DIRTY and the CLEAN cases show a robust spillover effect along the Park Range (Fig.6 a). This result is consistent with the results from

Saleeby et al. (2008). The difference in total precipitation seen between the CLEAN and the EMISS cases (shown in Fig.6 b) are much less. This is due in part to the fact that the aerosol concentrations in the EMISS case were much closer to the aerosol concentrations used in the CLEAN case (on average ~200 cm^{-3}). However, the spatial and temporal heterogeneities seen in the EMISS case caused an overall mitigating influence on the spillover effect. It was determined that polluted air upwind of small mountain barriers lead to a more localized spillover effect that replenished snow lost from spillover on the upwind side of the Park Range mountain barrier. The general conclusion of the orographic snow simulation is that the effect that CCN has on wintertime orographic precipitation is more complex than previous sensitivity studies suggested, owing to the spatial and temporal variations in CCN concentrations.

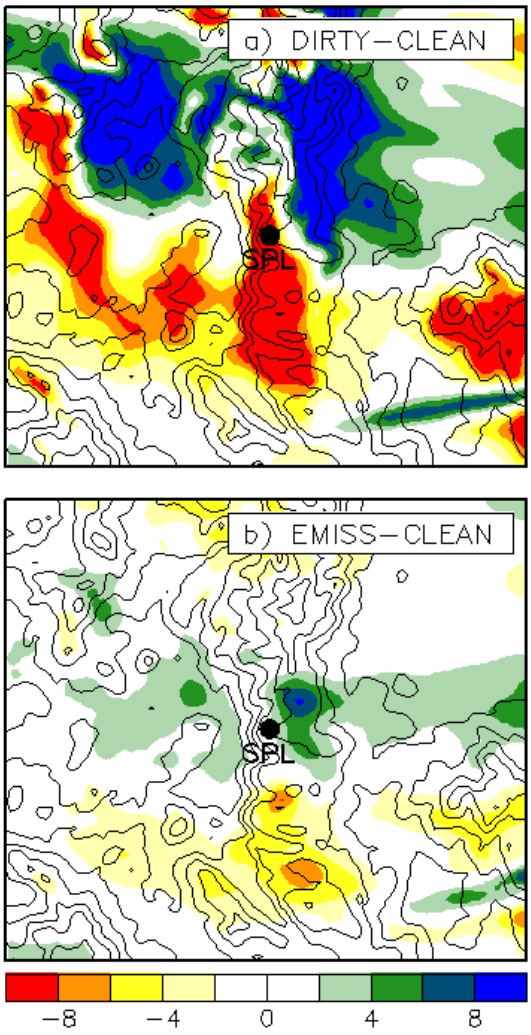

Illustration 6: a) Top: Total SWE differences between the DIRTY-CLEAN . b) Bottom: Total SWE differences between the EMISS-CLEAN

6. Conclusions

A simple and computationally efficient aerosol emissions parameterization was developed for RAMS in order to aid (primarily) in determining the effect of anthropogenic pollution on wintertime orographic precipitation. It was determined that this scheme was able to reasonably represent the temporal and spatial evolution of aerosol number concentrations despite a lack of secondary aerosol formation processes. Furthermore, when used as part of a the Feb 11th, 2007 snow event simulation, it was determined that the response of orographic clouds to CCN became more complex when spatially and temporally heterogeneous CCN concentrations were used.

7. Acknowledgments

This work was funded by the National Science Foundation through grants ATM 0835421 and AGS 1138896

8. References

Andrea, M.O., and D. Rosenfeld, 2008: Aerosol-cloud-precipitation interactions. Part 1. The nature and sources of cloud-active aerosols. *Earth. Sci. Rev.*, **89**, doi:10.1016/j.earscirev.2008.03.001.

Borys, R.D., D.H. Lowenthal, and D.L. Mitchell, 2000: The relationships among cloud microphysics, chemistry, and precipitation rate in cold mountain clouds. *Atmos. Env.*,**34**, 2593-2602.

Levin, Z., and W. R., Cotton, 2009: Aerosol pollution impact on precipitation, Springer, 386pp.

Liao, H., D.K. Henze, J.H. Seinfeld, S. Wu, and L.J. Mickley, 2007: Biogenic secondary organic aerosol over the United States: Comparison of climatological simulations with observations. *J. Geophys. Res.*,**112**, doi:10.1029/2006JD007813.

Lynn, B., A. Khain, D. Rosenfeld, and W. L. Woodley, 2007: Effects of precipitation from orographic clouds. *J. Geophys. Res.*, **112**, doi:10.1029/2006JD007537.

Petters, M.D., and S. M. Kreidenweis, 2007: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity. *Atmos. Chem. Phys*., **7**, 1962-1971.

Saleeby, S.M., W.R. Cotton, D. Lowenthal, R.D. Borys, and M.A. Wetzel, 2008: Influence of Cloud Condensation Nuclei on Orographic Snowfall. *J. Appl. Meteorol. Clim.,***48**, doi:10.1175/2008JAMC1989.1.

Saleeby, S.M., W.R. Cotton, and J.D. Fuller., 2011: The cumulative impact of cloud droplet nucleating aerosols on orographic snowfall in Colorado. *J. App. Meteo.***50**, 604-625.

Schichtel, B. A., W. C. Malm, G. Bench, et al. (2008), Fossil and contemporary fine particulate carbon fractions at 12 rural and urban sites in the United States, J. *Geophys. Res.-Atmos*., **113**.

U.S. Environmental Protection Agency, 2009: 2005 National Emissions Inventory Data and Documentation, http://www.epa.gov/ttnchie1/net/2005inventory.html.

Ward, D. and W. R. Cotton, 2011: A method for forecasting CCN using predictions of aerosol physical and chemical properties from WRF/Chem. *J. App. Met. Clim.*, **50**, 1601-1615.